IL-10670
S-95,247
Cust. No. 24981

METHOD OF FORMING A PACKAGE FOR MEMS-BASED FUEL CELL

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[0001] The United States Government has rights in this invention pursuant to Contract No. W-7405-ENG-48 between the United States Department of Energy and the University of California for the operation of Lawrence Livermore National Laboratory.

BACKGROUND OF THE INVENTION

[0002] Portable power sources of various types have been under development for many years. A serious need exists for portable power sources with significantly higher power density, longer operating lifetime, and lower cost. Present rechargeable and primary portable power sources have excessive weight, size, and cost with limited mission duration. For example, batteries covering power range from 1-200 Watts have specific energies ranging from 50-250 Whr/Kg, which represents two to three hours of operation for a variety of applications.

SUMMARY OF THE INVENTION

[0003] Aspects of the invention include a method comprising the steps of: patterning a package material into a preform layout; forming a package from the package material into a plurality of layers comprising at least a fuel reservoir interface layer, a layer containing a plurality of resistive heating elements, a microporous flow host structure layer containing a fuel cell, and a cap layer; and incorporating microchannels into the package.

[0004] Further aspects of the invention include a fuel cell package comprising: a first layer having a current input, a fuel inlet and a first plurality of electrical leads connected to the current input; a second layer having an anode manifold support structure, a fuel flow passage connecting to the fuel inlet and a fuel outlet; a third layer having a manifold support beam, a resistive heater support structure, a fuel flow passage, an air flow inlet connecting to an air flow passage, and a resistive heater connecting to each of the first plurality of electrical leads; a fourth layer having a fuel flow passage, an air flow passage, and a microporous flow host structure containing a thin film fuel cell formed from an electrolyte sandwiched between an anode and a cathode; a fifth layer having an air manifold connecting to the air flow passage in the fourth layer, a fuel flow passage, an anode electrical feedthrough, and a cathode electrical feedthrough; a sixth layer having an air flow passage connected to the air manifold in the fifth layer, a fuel flow passage, an anode electrical feedthrough and a cathode electrical feedthrough; and a seventh layer having an air flow passage, a fuel flow passage, an anode electrical feedthrough and a cathode electrical feedthrough; wherein , a resistive electrical feedthrough and an electrical feedthrough connected to a ground communicates through each of the layers.

[0005] Further aspects of the invention include a fuel cell package comprising: a first layer having a current input, a fuel inlet and a first plurality of electrical leads connected to the current input; a second layer having an anode manifold support structure, a fuel flow passage connecting to the fuel inlet and a fuel outlet; a third layer having a manifold support beam, a resistive heater support structure, a fuel flow passage, and a resistive heater connecting to each of the first plurality of electrical leads; a fourth layer having a fuel flow passage and a microporous flow host structure containing a thin film fuel cell

formed from an electrolyte sandwiched between an anode and a cathode; a fifth layer having an air containing means to allow air to breath into the fuel cell package, a fuel flow passage, an anode electrical feedthrough, and a cathode electrical feedthrough; a sixth layer, a fuel flow passage, an anode electrical feedthrough and a cathode electrical feedthrough; and a seventh layer having a fuel flow passage, an anode electrical feedthrough and a cathode electrical feedthrough; wherein , a resistive electrical feedthrough and an electrical feedthrough connected to a ground communicates through each of the layers.

BRIEF DESCRIPTION OF THE DRAWINGS

[0006] Figure 1A shows the first layer of a ceramic green tape fuel cell prior to assembly.

[0007] Figure 1B shows the second layer of a ceramic green tape fuel cell prior to assembly.

[0008] Figure 1C shows the third layer of a ceramic green tape fuel cell prior to assembly.

[0009] Figure 1D shows the fourth layer of a ceramic green tape fuel cell prior to assembly.

[0010] Figure 1E shows the fifth layer of a ceramic green tape fuel cell prior to assembly.

[0011] Figure 1F shows the sixth layer of a ceramic green tape fuel cell prior to assembly.

[0012] Figure 1G shows the seventh layer of a ceramic green tape fuel cell prior to assembly.

[0013] Figure 2 is an illustration of the third layer, the fuel manifold and resistive heater layer, of a ceramic green tape fuel cell prior to assembly..

[0014] Figure 3 is an illustration of a TFMPFHS (Thick Film Microporous Flow Host Structure) layer.

[0015] Figure 4A is cross-sectional view of a microfluidic fuel cell package showing air flow path.

[0016] Figure 4B is cross-sectional view of a microfluidic fuel cell package showing fuel flow path.

DETAILED DESCRIPTION

[0017] The invention herein describes a method of forming a package for a miniature fuel cell device. Illustrated in Figures 1A-1G is a preform layer of a seven layer fuel cell package. The package can be fabricated from a Low Temperature Co-fired Ceramic (LTCC), i.e., a ceramic green tape preform, such as Dupont 951 Green Tape, or a plastic or polymer preform, such as Dupont Kapton or Sylgard silicone. Methods to form the preform layers include laser cutting, injection molding, or extrusion molding of the ceramic or plastic.

[0018] Referring to Figure 1A, the first layer of the package, a fuel reservoir interface **2**, is fabricated from a ceramic green tape, molded ceramic, or a plastic preform. Fuel reservoir interface **2** comprises a resistive heater current input **4** having three electrical leads **6**, **8**, and **10**, a fuel flow passage **12**, a left side alignment pin **14**, a right side alignment pin **16**, and a grounded resistive heater feedthrough **18**. Fuel reservoir interface **2** may also comprise an airflow via **20** if forced air is used. Resistive heater

current input **4** can be connected to a load such as a battery or a super-capacitor, providing current flow to create heat in the resistor. Initial heating of the fuel cell package may also be introduced to the preform package through other techniques such as catalytically burning a hydrocarbon fuel-air mixture in a miniature isolated volume.

[0019] A second layer of the fuel cell package shown in Figure 1B is an anode manifold support and fuel/air flow through layer **21** that is fabricated from a ceramic green tape, molded ceramic, or a plastic preform. Anode manifold support and fuel/air flow through layer **21** comprises electrical feedthrough **5,** electrical leads **6**, **8**, and **10**, fuel flow passage **12**, left side alignment pin **14**, right side alignment pin **16**, and grounded resistive heater feedthrough **18**. Anode manifold support and fuel/air flow through layer **21** may further comprise airflow via **20** if forced air is used.

[0020] In Figure 1C, a third layer of the fuel cell package is a fuel/anode manifold and resistive heater layer **22** that is fabricated from a ceramic green tape, molded ceramic, or a plastic preform. Fuel/anode manifold and resistive heater layer **22** rests directly on top of the second layer, anode manifold support and fuel/air flow through layer **21**, allowing the electrical leads (**6,8,10**) to make continuous electrical contact to the first layer. Referring now to Figure 2, fuel manifold support and resistive heater layer **22** is shown in greater detail. This layer comprises electrical feedthrough **5**, left side alignment pin **14**, right side alignment pin **16**, fuel flow passage **12**, a resistive heater **24**, electrical leads **6, 8, 10** connected to electrical input **4** through electrical feedthrough **5**, manifold support beam **27** on which resistive heater **24** is formed, and three electrical leads **26** that are connected to ground through electrical feedthrough **18**. Fuel/anode manifold and resistive heater layer **22** provides the mechanical support for the next layer, which

includes the thick film microporous support structure. Additionally, the manifold support and resistive heater layer may optionally contain an air flow via **20** if forced air is used. Resistive heaters are formed along the top surface of the manifold support beam **27**. The heaters are connected to a common feedthrough electrical lead that is grounded at one end, and at the other end to feedthroughs connected to a common input electrical feedthrough. The input feedthrough can be connected to a small battery that can power the heater. Manifold support beam **27** and resistive heater layer **22** provide physical support beams which support microporous flow host structure.

[0021] Referring Figure 1D, a Thick Film Microporous Flow Host Structure (TFMPFHS) layer **28** forms the fourth layer of the fuel cell package. TFMPFHS layer **28** comprises electrical feedthrough **5**, a microporous flow host structure (not shown), a fuel flow passage **12**, left side alignment pin **14**, right side alignment pin **16**, and grounded resistive heater feedthrough **18**. TFMPFHS layer **28** may further comprise an airflow via **20** if forced air is used. TFMPFHS layer **28** forms a drop-in template **30** where a thick film microporous flow host structure (not shown) is positioned. Additional approaches can form the TFMPHS in a continuous ceramic laminate structure rather than forming a drop-in template. For this embodiment, the entirety of TFMPFHS layer **28** in Fig 1 can be a thin film fuel cell formed on a ceramic or plastic laminate layer having a plurality of pores. The laminate can further include fuel and air feedthroughs as shown in Fig 1, and the thin film fuel cell can be patterned in an appropriate template on the porous laminate layer so as to cover the center regions of pores, but not extend to the areas where the fuel and air flow channels are located.

[0022] TFMPFHS layer **28** contains a thin film fuel cell at its top surface. Effective fuel cells are described elsewhere in pending U.S. Patent application S-88,911 which is hereby incorporated by reference. Referring to Figure 3, a microporous flow host structure **31** comprises a thin film fuel cell **32**, an anode contact **34**, and a cathode contact **36**. The fuel cell (not shown) includes a porous anode/catalyst layer, a dense electrolyte layer, and a porous cathode layer. The fuel cell can be either a proton exchange membrane (PEM) or solid oxide fuel cell (SOFC) materials structure. For the PEM fuel cell, the anode can be a thin film of nickel or carbon on the porous host structure, followed by a platinum or platinum-ruthenium catalyst. This is followed by the electrolyte material, which can be Nafion. The cathode can have a platinum catalyst, followed by another carbon or Ni porous electrode. PEM fuel cells typically operate at temperatures between about 60°C and about 90°C. Similarly, an SOFC structure can be formed by depositing a Ni anode on the porous host structure, followed by an anode catalyst, such as cerium oxide (CeO_2), after which is located a dense electrolyte layer, such as yttria-stablized zirconia (YSZ). The dense electrolyte layer is followed by a cathode catalyst, such as cerium oxide (CeO_2), which is then followed by a porous electrode material, such as silver or lanthanum strontium manganate. In the invention, fuel is allowed to flow between the support beams, allowing fuel to come into contact with a majority of the anode surface area by way of the micropore passages in the thick film host structure.

[0023] Figure 1E illustrates an air manifold layer **38** that forms the fifth layer of the fuel cell package. Air manifold layer **38** comprises electrical feedthrough **5**, an air manifold **40**, left side alignment pin **14**, right side alignment pin **16**, an anode electrical

feedthrough **42**, a cathode electrical feedthrough **44** fuel flow passage **12**, and air flow via **20** if forced air is used.

[0024] Figure 1F illustrates an air manifold support layer **54** that forms the sixth layer of the fuel cell package. Air manifold support layer **54** comprises electrical feedthrough **5**, an air flow via **20** communicating with the air manifold **56**, left side alignment pin **14**, right side alignment pin **16**, anode electrical feedthrough **42**, cathode electrical feedthrough **44** and fuel flow passage **12**.

[0025] Figure 1G illustrates a ceramic green tape or Plastic preform cap **46** that forms the seventh layer of the fuel cell package. Cap **46** aligns directly over air manifold layer **38** forming sealed bonds around electrical feedthroughs **5, 42, 44**, fuel flow passage **12** and air flow via **20**. Cap **46** can serve as the final layer in the fuel cell package. If the fuel cell package contains several fuel cells, cap **46** acts as a common layer, i.e., serving the dual function of capping off a first sub-package while simultaneously serving as a microfluidic interface and support structure for a second sub-package (not shown). Cap **46** comprises electrical feedthrough **5**, left side alignment pin **14**, right side alignment pin **16**, anode electrical feedthrough **42**, cathode electrical feedthrough **44**. If connected to a second sub-package (not shown), Cap **46** contains airflow via **20** and fuel flow passage **12**.

[0026] Figure 4A shows a cross-sectional view of the first seven layers of a fuel cell package **48** showing the fuel flow path **52** in a cross flow configuration. Figure 4B shows the first seven layers of a fuel cell package showing the air flow path **50** and the fuel flow path **52** in a cross flow configuration. Inlet flow passages for fuel, i.e. **52**, and oxidant (air) sources, i.e. **50**, are provided in the first layer which enable direct interface

and design of heat transfer characteristics between the fuel cell package and a fuel storage reservoir (not shown) usually connected to the microfluidic fuel inlet using a valve, microvalve or other interconnect scheme.

[0027] Air flow **50** and air flow via **20** facilitate the use of forced air through the fuel cell package. Forced air is not necessary if an air "breathing" system is used. An air breathing system, for example, can contain perforations within the air manifold layer **38** that extends to the exterior of the package structure acting as a series of conduit that effectively provides air to the fuel cell.

[0028] The package material can comprise either a molded plastic or a ceramic green tape material. These materials are available in various thicknesses ranging from about 25μm to about 1 mm (typically ranging from about 50μm to about 250 μm) and can be shaped and patterned into arbitrary perform layouts using various etch or molding techniques. Etch techniques can, for example, include laser machining, wet etch or plasma etch. Extrusion molding and injection molding are examples of effective molding techniques. Metal interconnects can be patterned on these materials by any conventional means such as using screen print techniques.

[0029] A benefit of using ceramic green tapes for fuel cells is that the ceramic materials can be tailored to provide either high thermal conduction or high thermal isolation. This tailoring allows, for example, the center of the package to be concentrated at a high temperature while keeping the outer area cool, i.e., the operating temperature of the fuel cell can be between about 300°C to about 650°C while the fuel cell package remains cool enough to handle with a bare hand, i.e. less than about 55°C. Specific microfluidic cooling designs can be included in the laminated preform designs to provide counterflow

heat exchange, thereby heating incoming cool gases with exhausted hot gas streams. Another benefit of using ceramic green tapes is that the ceramic preforms can have metal feedthroughs that enable electrical contact to conductive lead materials such as metal leads which can be made of, for example, silver or Platinum. The metal feedthroughs can extend vertically between the layers of the ceramic tape layers allowing several fuel cells to be stacked together in a three-dimensional layout. Another advantage for using ceramic green tapes is that resistive heating elements controlling the temperature of the electrode-electrolyte-electrode layers, i.e., the fuel cell stack, can be incorporated into the package. Additionally, microchannels that allow delivery of liquid fuel, and oxidant to specific sides of the fuel cell stack can also be incorporated into the package if ceramic green tape materials are used. In this embodiment, the inlet fuel passages can be coated with catalyst materials, such as Pt, Pt-Ru, Ni, or Cu-ZnO, which when heated assist in converting a liquid hydrocarbon fuel to hydrogen and other byproducts.

[0030] The microporous flow host structure can be silicon, ceramic, anodic alumina, plastic, or other similar material that contains a high density of porous flow channels formed therethrough, which allows direct flow of fuel to the porous anode structure of the fuel cell. The anode and cathode electrodes are patterned such that interconnect pads are positioned where they can make electrical contact to feedthroughs connected to the exterior of the package or the adjacent fuel cell positioned in the package.

[0031] Air manifold layer **38** provides the electrical feedthroughs for the anode, cathode, and resistor power input, as well as fuel and oxidant flow channels if necessary to connect to the adjacent level fuel cell in the stack. Air manifold layer **38** further provides a manifold to distribute the air to the cathode structure. In addition, air manifold layer **38**

acts as a sealing means, such as, an o-ring seal around the top periphery of the microporous flow host structure that was inserted into TFMPFHS layer **28**. A thin preform of Kapton tape or silicon dioxide tape can also be used to form a sealing bond beneath air manifold layer **38**, or the forming properties of the plastic or ceramic green tape layers can be exploited to both bond and seal the microporous flow host structure / thin-film fuel cell into the package. Preferred methods and materials will depend on the desired operating temperature of the fuel cell package.

[0032] Ceramic green tape or plastic preform cap **46** is similar to the original sub-package microfluidic interface, except cap **46** contains electrical feedthroughs that enable simple fexibility when stacking and scaling the total number of fuel cells in the package.

[0033] The package is formed by aligning and contacting the package material layers. For instance, a green tape material contains a plastic binder materials which holds the thin sheets in form. The green tape structure is cofired in a furnace which removes the plastic binder and also forms a bond between the layers to thus, permanently connect the layers. Microporous flow host structure **30** is inserted within the layers as shown in Figure 1. If any of the components of the microporous flow host structure cannot withstand the firing temperature of the ceramic tape, then the preform layers can be co-fired, i.e., all layers baked simultaneously, and assembled with the fuel cell using a low temperature adhesive to form the final bond and seal.

[0034] While particular operational sequences, materials, temperatures, parameters, and particular embodiments have been described and or illustrated, such are not intended to be limiting. Modifications and changes may become apparent to those skilled in the art, and it is intended that the invention be limited only by the scope of the appended claims.